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# RAYLEIGH LIGHT SCATTERING FOR CONCENTRATION MEASUREMENTS IN TURBULENT FLOWS

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## INTRODUCTION

Despite intensive research efforts over a number of years, an understanding of scalar mixing in turbulent flows remains elusive. An understanding is required because turbulent mixing has a pivotal role in a wide variety of natural and technologically important processes. As an example, the mixing and transport of pollutants in the atmosphere and bodies of water are often dependent on turbulent mixing processes. Turbulent mixing is also central to turbulent combustion which underlies most hydrocarbon energy utilization in modern societies as well as unwanted fire behavior. Development of models for combusting flows is crucial for more efficient utilization of limited hydrocarbon fuel resources, reduction in environmentally harmful pollutants, more efficient chemical processes, and for the prediction of hazard associated with unwanted fire. However, an understanding of scalar mixing is required before useful models of turbulent mixing and, ultimately, turbulent combustion can be developed.

An important subset of turbulent flows is axisymmetric turbulent jets and plumes because they are relatively simple to generate, and because they provide an appropriate test bed for the development of general theories of turbulent mixing which can be applied to more complex geometries and flows.

During this talk we will focus on a number of experimental techniques which have been developed at NIST for measuring concentration in binary (i.e., consisting of two components) axisymmetric turbulent jets. In order to give a flavor for the value of these diagnostics, some of the more important results from earlier and on-going investigations will be summarized. Topics to be addressed include the similarity behavior of variable-density axisymmetric jets [1],[2], the behavior of absolutely unstable axisymmetric helium jets [3], and the role of large-scale structures and scalar dissipation in these flows [4],[5].

## RAYLEIGH LIGHT SCATTERING FOR CONCENTRATION MEASUREMENT

Rayleigh light scattering refers to the elastic scattering (i.e., no change from the incident wavelength) of light by molecular species. For a mixture of  $m$  gases, the intensity of RLS is given by

$$I(90^\circ) = N \left( \sum_{j=1}^m \sigma_j(90^\circ) X_j \right) I_o \quad (1)$$

where  $N$  is the total number density of molecules,  $\sigma_j$  is the Rayleigh scattering cross section for the wavelength of incident light,  $X_j$  is the mole fraction of species  $j$ , and  $I_o$  is the incident light irradiance. This expression assumes that the molecules are isotropic. For this case the scattered light has the same polarization as the incident light. More complicated expressions are required for scattering from anisotropic molecules which depolarize the scattered light. [6]

A typical RLS experiment involves monitoring the scattered light intensity from a short length of a laser beam located within a gas flow. The information which can be determined from the signal depends on the type of gas which is located within the observation volume. For isothermal binary mixtures it is typical to perform calibrations of the RLS scattering intensities for the two unmixed gases. The resulting expression for calculating the mole fraction of gas 2,  $X_2(t)$  where the  $t$  dependence indicates that  $X_2$  may fluctuate with time, is quite simple,

$$X_2(t) = \frac{I(t) - I_1}{I_2 - I_1} \quad (2)$$

Here  $I(t)$  is the time-dependent intensity of RLS from the volume containing the gas mixture of interest, and  $I_1$  and  $I_2$  are observed RLS intensities for scattering from pure gases 1 and 2. The mole fraction of gas 1 is simply obtained from Eq. (2) as  $X_1(t) = 1 - X_2(t)$ . Once mole fractions are available, mass fractions,  $Y_1(t)$  and  $Y_2(t)$ , can be calculated using standard formulas. An important advantage of Eq. (3) is that only relative values of RLS intensity need to be recorded. It is not necessary to measure absolute intensities. This greatly simplifies the optical system and analysis requirements.

An important limitation of RLS is that resonant scattering from small particles (Mie scattering), which are generally present in laboratory environments, and surfaces (glare) can be much stronger than RLS. These light sources act as interferents and noise sources and, in worst cases, can preclude RLS measurements.

## RLS MEASUREMENT OF CONCENTRATION IN TURBULENT FLOW FIELDS

By using either pulsed (e.g., [7]) or continuous wave (cw, e.g., [8]) high-powered lasers to induce RLS, it is possible to measure concentration in turbulent flow fields of two gases with high temporal and spatial resolution. Pulsed lasers are usually employed to make multipoint (line or planar) measurements. These lasers generally generate very short pulses (in the ns to  $\mu$ s range) which "freeze" the turbulent motion, but have low repetition rates (typically 10 Hz) which preclude following the time behavior of the flow. cw lasers have usually been used for point measurements, but measurements along a line defined by a laser beam have also been demonstrated. [9] Data rates for such measurements are generally high enough to allow real-time recordings of RLS scattering to resolve the turbulent concentration fluctuations within a flow.

## EXPERIMENTAL SYSTEMS DEVELOPED AT NIST

### FLOW SYSTEMS

Over the years a series of flow systems and experimental configurations have been developed at NIST for the measurement of concentration in turbulent flow fields generated by the flow of axisymmetric jets into a second gas. The focus has been on the development of systems capable of real-time measurements having high spatial and temporal resolution. Only brief descriptions of the experimental systems are provided here. Interested readers are referred to the cited references for additional details.

Two different approaches have been used to overcome the problem of Mie scattering from small particles interfering with RLS measurements of concentration. In our initial experiments a coflow system was used in which an axisymmetric gas jet from a 6.35 mm pipe entered a slow coflow of a second gas contained within a square glass enclosure having 10 cm sides. [1],[6],[10] Both flows were filtered to remove particles. Most turbulent studies using RLS have used similar configurations to limit Mie scattering interferences since accurate measurements have proven difficult in the presence of particles. [11]

Recently a new experimental system, termed the Rayleigh Light Scattering Facility, has been developed to provide a large enough dust-free volume to allow measurements of free jets. [12] A flow of air filtered by high efficiency particle filters is used to sweep particles from within a 2.3 m long by 2.3 m diameter cylindrical test section. Experiments are performed by turning off the blowers and waiting for flow transients to dissipate. This system provides a nearly ideal environment for RLS studies of mixing since both Mie scattering and glare are barely detectable.

Jets within the enclosure have been formed either from the pipe flow described above or by using a contoured nozzle designed to generate a top hat velocity profile at the jet exit. The enclosure includes a three-dimensional positioning system for orienting the flow field relative to the optics which are fixed within the enclosure.

### SINGLE-POINT MEASUREMENTS OF CONCENTRATION

The use of RLS scattering for concentration measurements in turbulent flows has been discussed in detail by Pitts and Kashiwagi. [6] The optical system required for single-point RLS measurements is fairly simple. It consists of a lens to focus the laser beam from an 18 W Ar ion laser to a narrow waist (50  $\mu\text{m}$  is a typical diameter) and an  $f/2$  light collection system for imaging the scattered RLS at right angles onto a pinhole which defines the length of the observation volume. Light is then detected by a photomultiplier tube for which the current output (proportional to detected light intensity) is converted to a voltage, digitized, and stored in the memory of a computer for later analysis. Data sets consisting of several hundred thousand measurements can be recorded.

Since RLS is a relatively weak optical process, the largest noise source is usually the Poisson statistics associated with photon detection, which is equal to the square root of the number of detected photons. [13] The intensity of RLS varies with the gas pairs studied so the noise level for a given experiment depends on the gases in the mixture. However, it has been shown that the current experimental arrangement allows measurements of real-time concentration having accuracies better than 1% of full scale at data rates of several thousand Hz for observation volumes as small as 0.0003  $\text{mm}^3$ . [6] By careful design of the optical system, Dowling and Dimotakis have achieved somewhat higher temporal and spatial resolution. [14]

### REAL-TIME LINE MEASUREMENTS OF CONCENTRATION

Single-point measurements are recorded by using optics to isolate a short length of scattered laser light from a laser beam. Clearly, if it were possible to rapidly image several individual lengths of the laser beam onto a number of detectors it would be possible to record real-time concentration fluctuations at a number of points simultaneously. A line camera system has been developed which is capable of recording propane (which has a particularly strong RLS signal) mole fraction at 128 adjacent points with a maximum line data rate of 2.37 kHz. The camera design required the incorporation of an image intensifier. Due to the relatively high noise level associated with the line scanner used in the camera and the desire for high temporal response, it was necessary to use a generation I image intensifier equipped with a high-speed phosphor. An earlier version of the line camera has been described in detail. [9] It incorporated a two-stage, generation I image intensifier equipped with a P-46 phosphor. A later version of the camera having a thirteen-fold improvement in signal-to-noise ratio utilizes a three-stage image intensifier with a P-47 phosphor screen. [15]

The line camera is designed for 1:1 focusing of a 14.7 mm length of the laser beam onto the image intensifier. A 4:1 fiber optic taper couples the output of the image intensifier to the 3.2 mm length of the 128 pixel line scanner. The effective spatial resolution is  $\approx 0.2$  mm. The line scanner limits the maximum line read-out

rate to 2.37 kHz. The output of the line scanner is digitized and stored by a computer. Many thousands of line scans can be recorded during a single experiment.

## COMBINED INSTANTANEOUS TWO-DIMENSIONAL AND REAL-TIME LINE MEASUREMENTS OF CONCENTRATION

Recently, Richards and Pitts have recorded two-dimensional images of RLS using a cooled CCD array to image RLS from a sheet generated by passing the second harmonic (532 nm wavelength) of a  $\text{Nd}^{+3}/\text{YAG}$  laser through a series of cylindrical lenses. [5] This experimental approach was first used by Escoda and Long. [7] Since the laser generates a 10 ns pulse, the flow field is "frozen" and two-dimensional images of concentration can be derived.

In addition to the sheet from the YAG laser, the cw Ar ion laser beam was passed through the flow field such that it was aligned parallel to and just downstream of the laser sheet. The real-time line camera was used to record measurements of concentration at this position. In this way it was possible to monitor the concentration fluctuations generated by a flow whose two-dimensional distribution was recorded just upstream an instant earlier.

## RESULTS

### SIMILARITY BEHAVIOR OF VARIABLE-DENSITY FLOWS

It has been known for some time that constant-density turbulent axisymmetric jets develop a self-similar behavior in which only a single length scale is necessary to characterize the time-averaged and fluctuation concentration profiles. (e.g., [16]) An important question was whether or not jets having global density variations due to density differences between the jet and surroundings would also develop self-similar behavior with increasing downstream distance as their densities approached that for the ambient gas due to continual entrainment of surrounding fluid into the jet.

Earlier experiments (including work from our laboratory) indicated that such flows did not achieve self-similar behavior. [1],[17] The Rayleigh light scattering facility has allowed careful measurements of the scalar and fluctuation fields for jets of helium, methane, and propane entering ambient air for which buoyancy effects were unimportant. [2] The findings of this investigation show that these flow fields do achieve full self-similar behavior. Profiles for concentration fluctuations also obey similarity relationships. The similarity profiles agree well with those measured for constant density jets by Dowling and Dimotakis. [14]

Propane jet measurements were recorded for both pipe and contoured-nozzle flows, which are expected to have quite different initial velocity profiles and turbulence levels. The results indicate that the final similarity state achieved by the variable-density flows is independent of the initial density ratio as well as the velocity distribution.

Very recently the single-point RLS technique has been used to characterize the mixing behavior in the near-field of helium jets from a contoured nozzle which are known to be absolutely unstable. Absolutely unstable jets develop intense and highly coherent oscillations in their near-field shear layers. [18] The strength of the effect results in a number of interesting vortical interactions which can lead to the formation of "side jets", a process in which jet fluid is vigorously ejected into the surrounding ambient fluid. As a result of side-jet formation, the near-field mixing rate is substantially enhanced compared to axisymmetric jets which do not display this behavior.

The RLS scattering measurements have been used to characterize mixing processes in the jet shear layer as well as in the side jets outside of the shear layer. These measurements have identified the parameters which control the strength of the absolute instability, provided insights into the vortical interactions which ultimately lead to the formation of side jets, and demonstrated that the strength of side jets is proportional to the strength of the instability in the shear layer. The side jets themselves have been shown to consist of nearly pure jet fluid, which indicates that they must come from the jet core. They are highly localized in space and tend to rotate randomly about the parent jet.

## ORGANIZED MOTION AND SCALAR DISSIPATION IN AXISYMMETRIC JETS

The line camera has been used to record real-time line images of concentration for a propane jet flowing from a nozzle into quiescent air. Measurements were made for various radial sections located 40 radii downstream from the jet exit. Three thousand line scans (384,000 individual concentration measurements) were recorded during a single experiment. The availability of such large data sets allows a number of statistical properties for the concentration field to be calculated. The time-averaged radial mass fraction profile is found to be in excellent agreement with earlier measurements in the same flow system. [2]

These real-time line measurements reveal that strong mixing occurs near the centerline. As one moves further from the centerline, the flow becomes intermittent (both mixed jet fluid and ambient air are observed). The time structure found for the turbulent fluid in this region is quite distinctive. For a particular radial location, it is observed that when the turbulent fluid first appears (i.e., at the end of a time period during which only air is present) there is a very rapid increase in jet fluid concentration. This rapid increase is followed by much slower fluctuations in concentration which gradually drop off until ambient air is once again observed. Such a behavior has often been noted in single-point scalar measurements and has been referred to as "ramp-like" structures. The line images demonstrate that ramp-like structures extend across a large radial extent of the flow. This is a clear indication that the motions which generate large-scale turbulent structures (LSTS) are organized. Previous measurements suggest that the downstream edges of the structures are the result of strong ejections of fluid from the central region of the jet, and that air entrainment occurs in upstream regions of the LSTS. [19]

The line camera results have also been used to estimate values for the time-resolved component of scalar dissipation in the radial direction ( $\chi_r$ ). Scalar dissipation, defined as

$$\chi = 2D\nabla Y \cdot \nabla Y, \quad (3)$$

where  $D$  is the binary molecular diffusion coefficient and  $Y$  is the mass fraction of jet fluid, is a measure of molecular mixing rate in turbulent flows. Values of  $\chi$  play a central role in the modeling of turbulent mixing in both isothermal and combustor turbulent flows.

The space-time images of  $\chi_r$  show that rapid spatial variations in the rate of mixing occur over small regions of space in the interior regions of the jet, but that large contiguous regions of rapid mixing are associated with large-scale structures on the outer edges of these flows.

The last set of results to be discussed are preliminary measurements in which instantaneous two-dimensional images of propane mole fraction have been recorded simultaneously with real-time line measurements at a location just downstream of the sheet location. The purpose of these measurements is to gain insight as to how the structures observed in the line measurements are related to the turbulent structures observed in instantaneous two-dimensional images and how LSTS are modified as they convect downstream.

## CONCLUSIONS

A number of experimental diagnostics for characterizing scalar fields in turbulent flows of two gases have been developed at NIST. Unique capabilities include experimental systems which allows measurements of free shear flows and a camera capable of recording real-time concentration at 128 points along a line simultaneously. These techniques have been used to 1) demonstrate that variable density jets obey similarity behavior, 2) characterize the boundary-layer structure and side jets in absolutely unstable axisymmetric jets, 3) demonstrate that LSTS play a fundamental role in the mixing behavior of axisymmetric jets, and 4) provide an improved understanding of scalar dissipation and its dependence on LSTS. Results of these studies along with those from other laboratories will ultimately provide the understanding necessary to develop effective models for predicting mixing behaviors of complicated turbulent flows and combustion systems.

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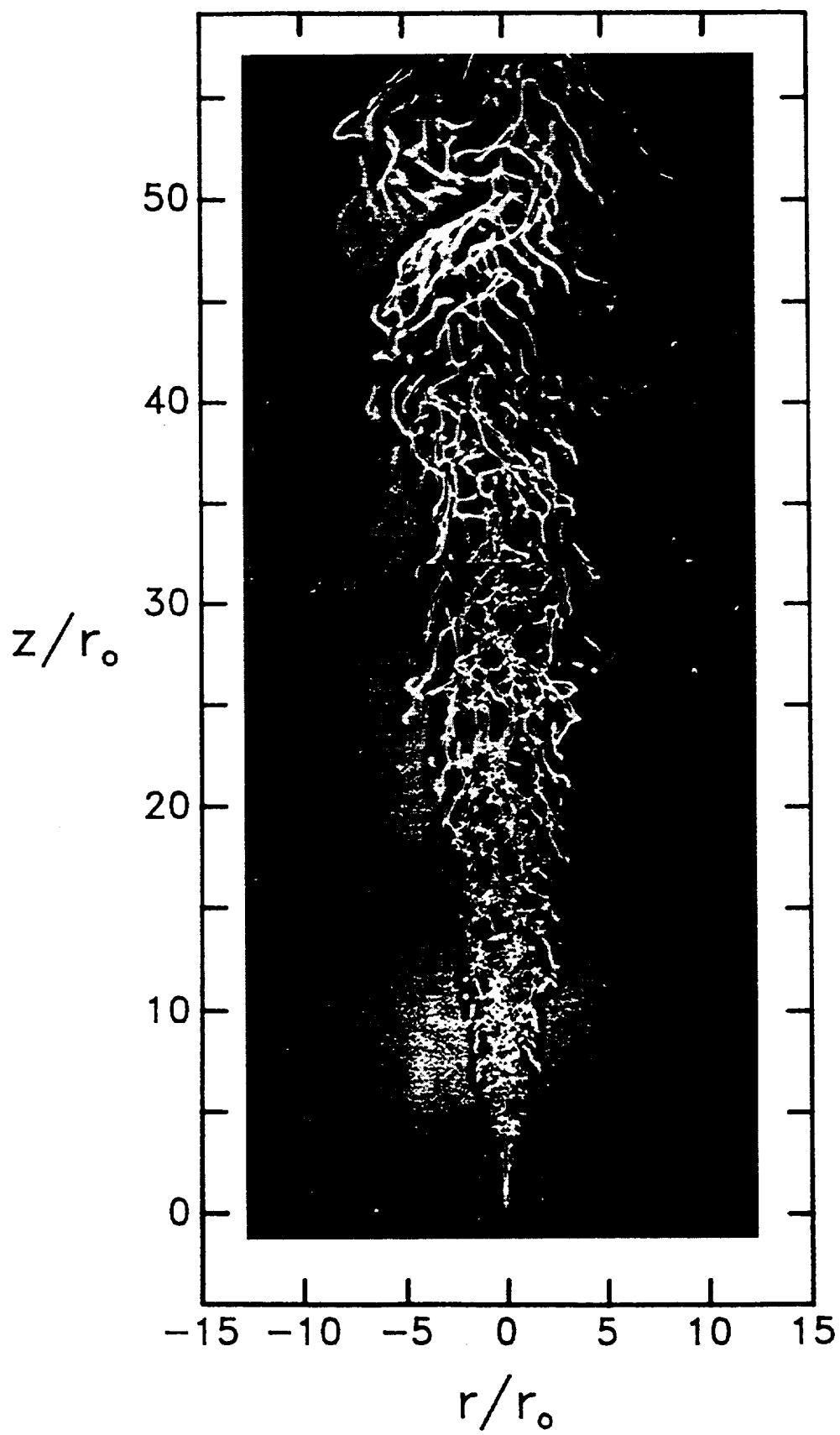
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## OUTLINE

1. Introduction to turbulent jets and real-time Raleigh light scattering measurements of concentration in binary turbulent jets
2. Rayleigh Light Scattering Facility
3. Point measurements in variable density jets
4. Point measurements in absolutely unstable helium jets
5. Real-time line camera for measurements of concentration
6. Line-camera results in turbulent propane jet
7. Combined real-time line camera and instantaneous two-dimensional measurements of concentration in turbulent propane jet
8. Summary

$\text{C}_3\text{H}_8/\text{air}$ ,  $\text{Re} = 3960$





## RAYLEIGH LIGHT SCATTERING

$$R_p = I_L \eta_L l \Omega \eta_c \epsilon N \sum_{j=1}^M \sigma_j X_j$$

$$R_p = KN \sum_{j=1}^M \sigma_j X_j$$

For Binary Gas Mixture:

$$X_2(t) = \frac{I(t) - I_1}{I_2 - I_1}$$

with  $I_1$  and  $I_2$  equal to the scattering from the pure gases 1 and 2

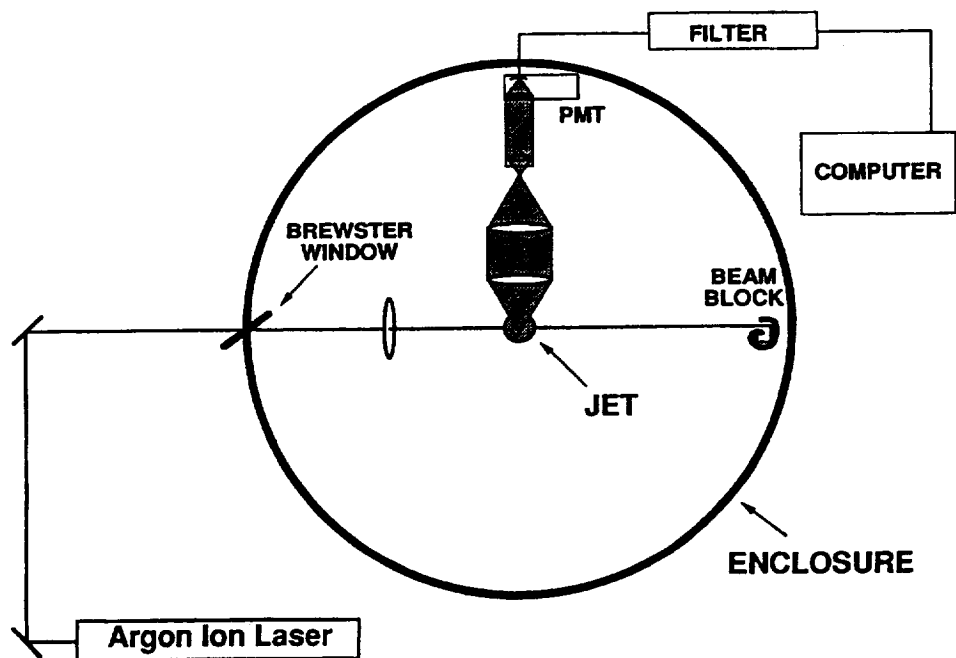
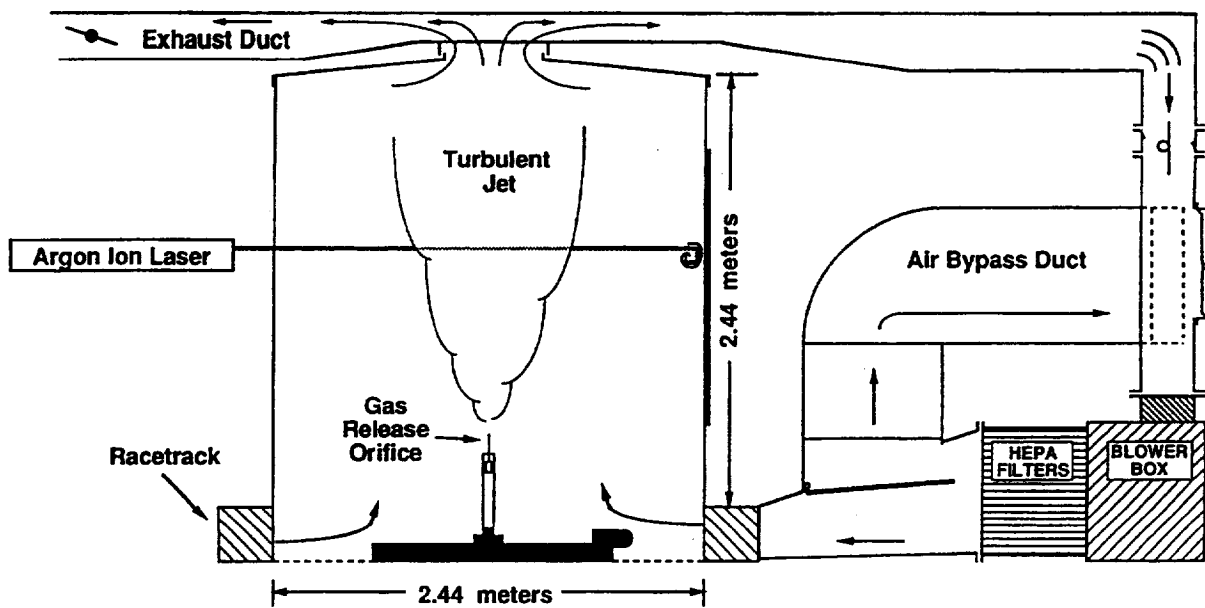
## RAYLEIGH LIGHT SCATTERING

### Advantages:

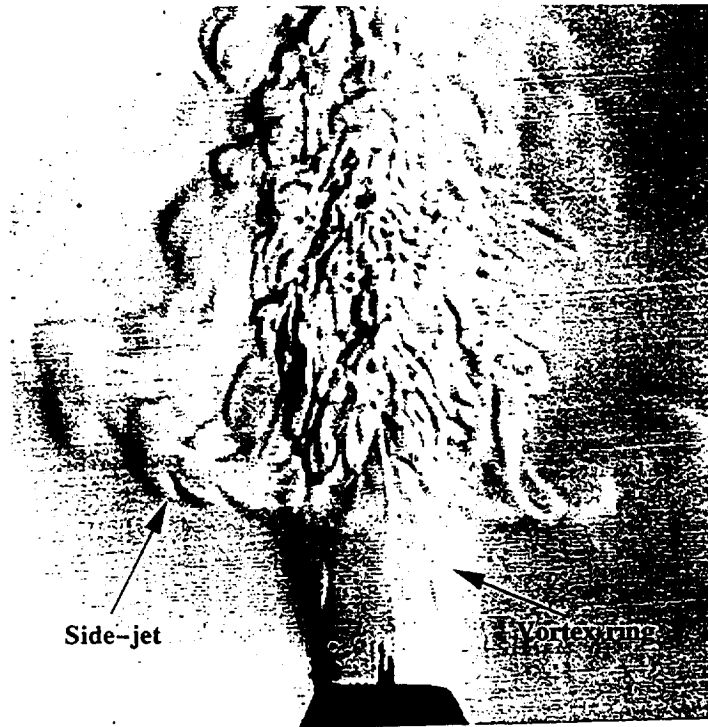
- Relatively strong signal allows real-time measurements
- Signal due to gas composition

### Disadvantages:

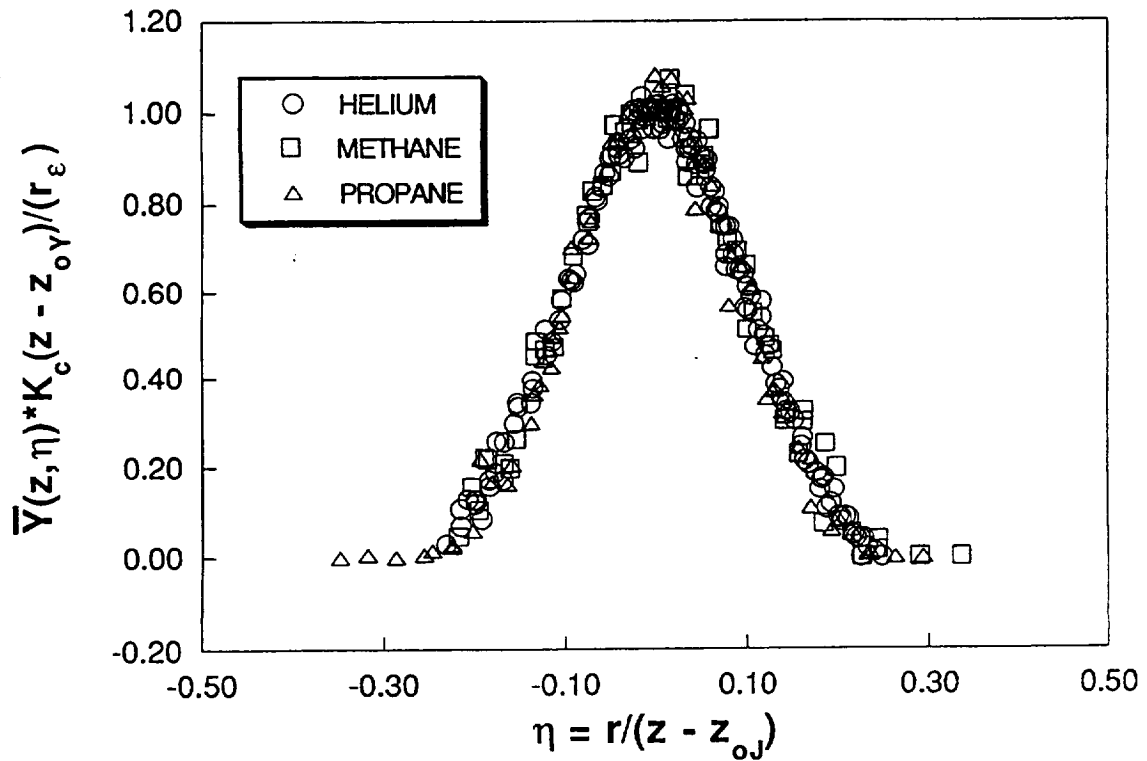
- Scattering occurs from all molecules
- Light is elastically scattered
  - glare
  - Mie scattering



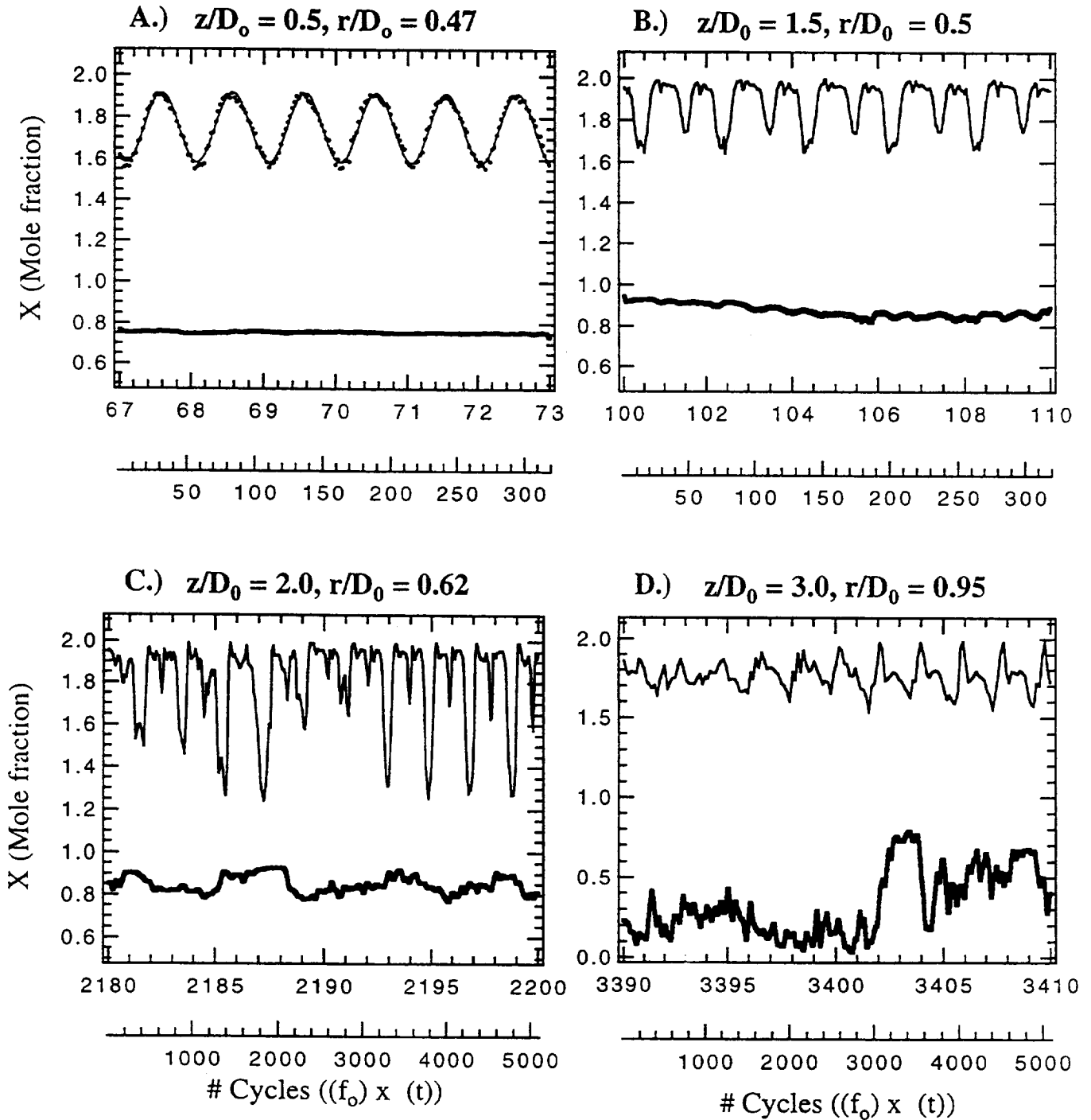
# GLOBALY UNSTABLE JET PRODUCING SIDE-JETS



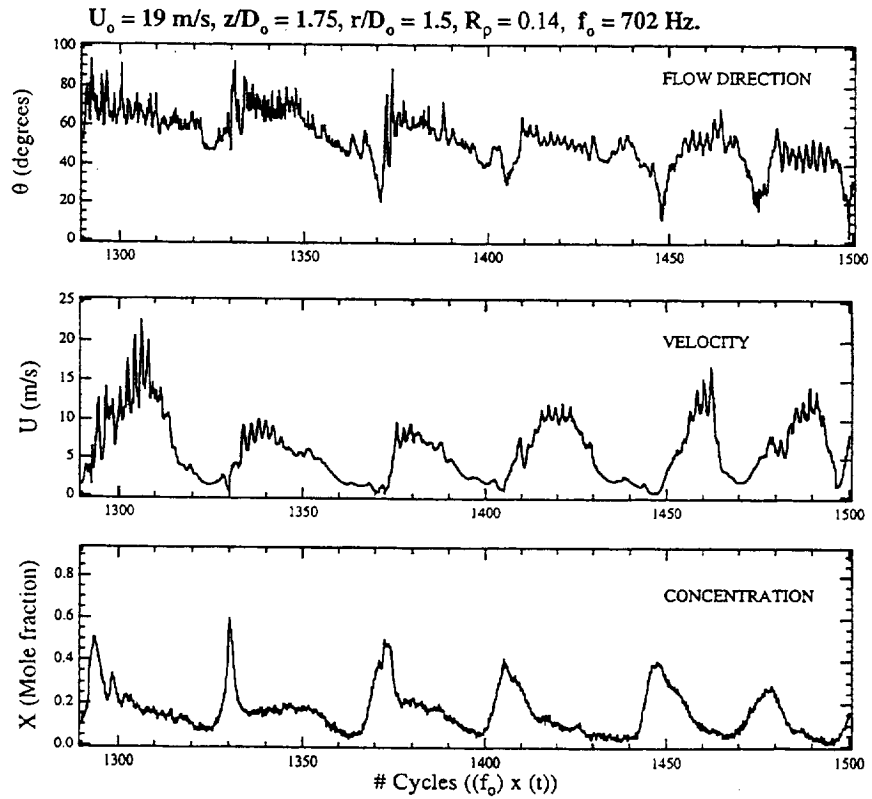
## FULL SIMILARITY, ALL GASES



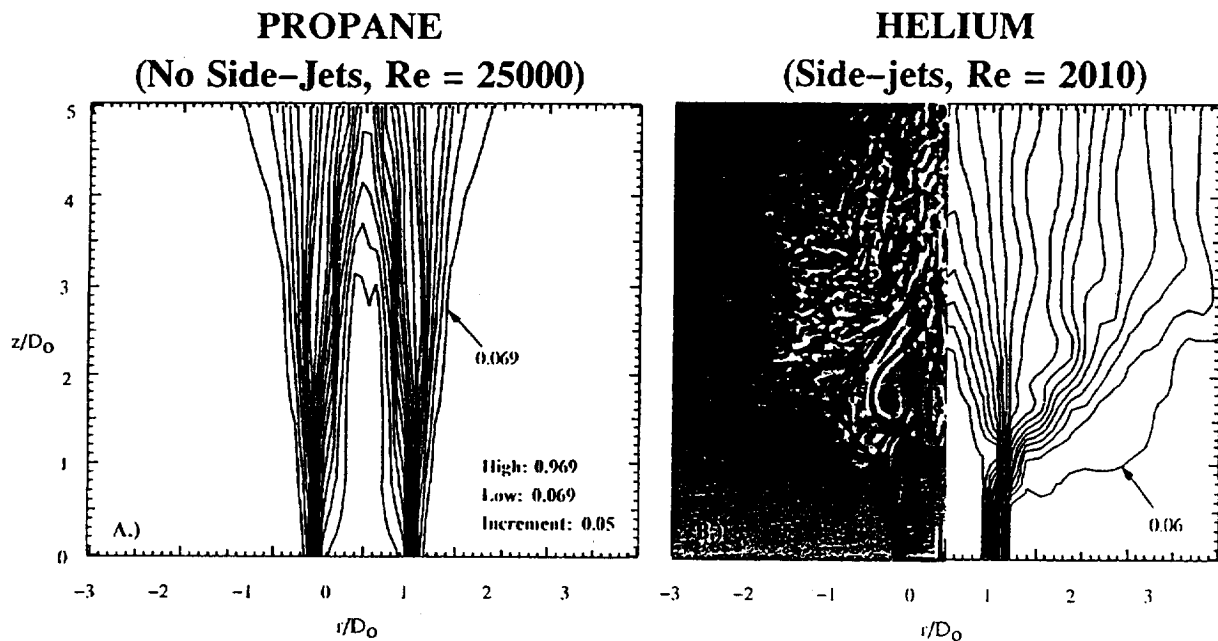
# **CONCENTRATION TIME-TRACES** **(Axial direction, shear layer)**



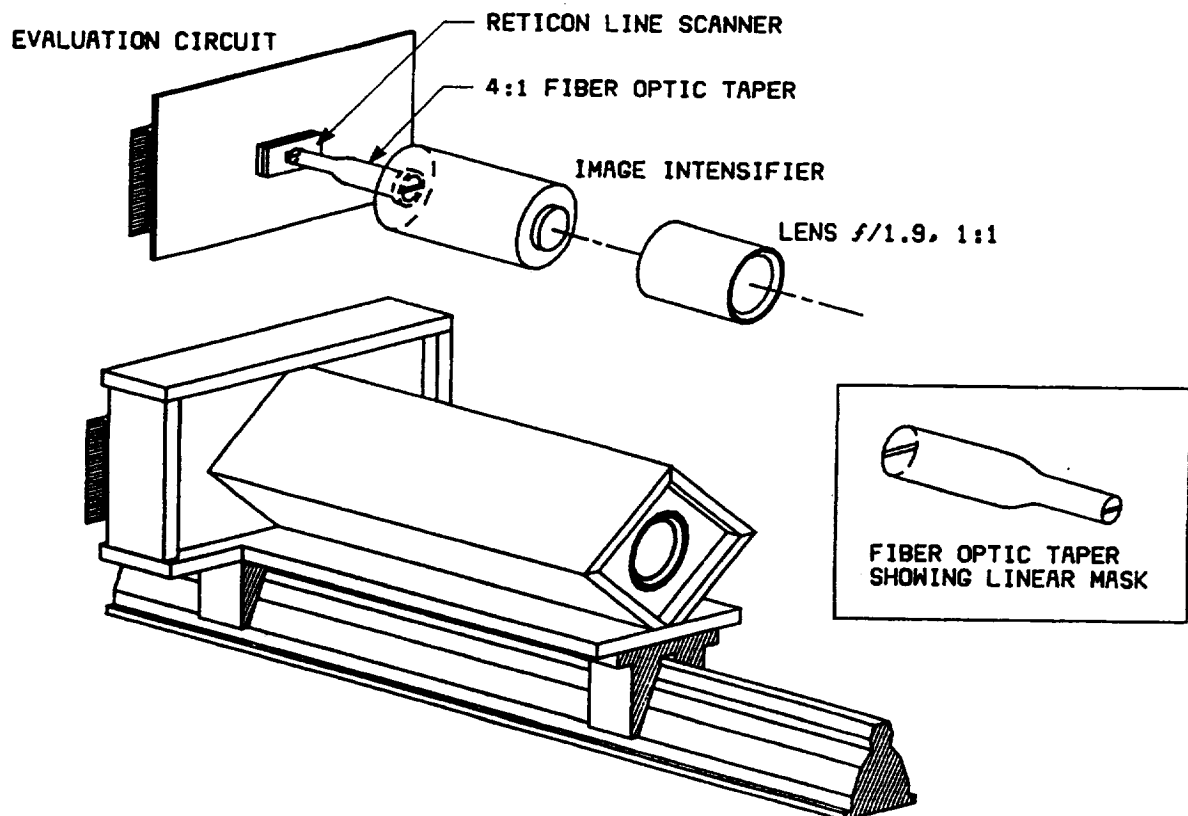
### Simultaneous velocity, concentration, angle (Side-jet fluid)

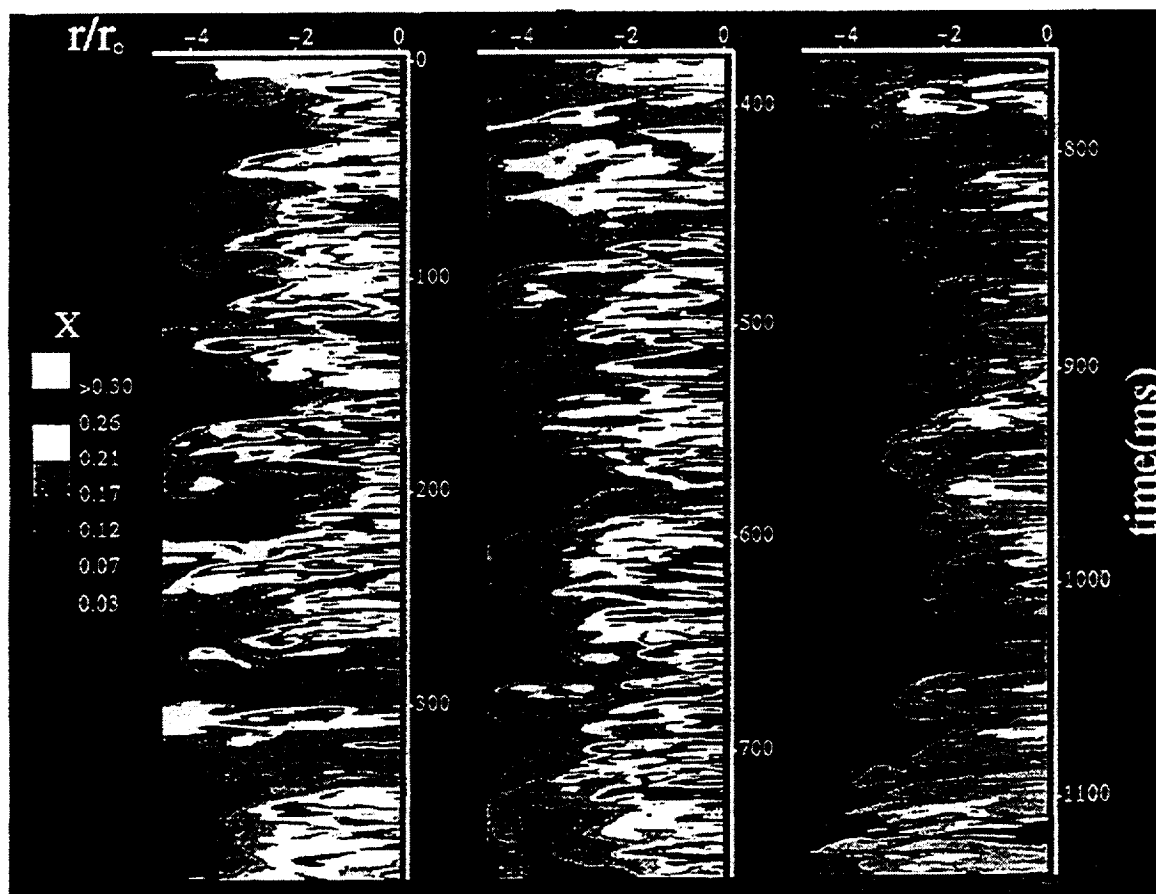
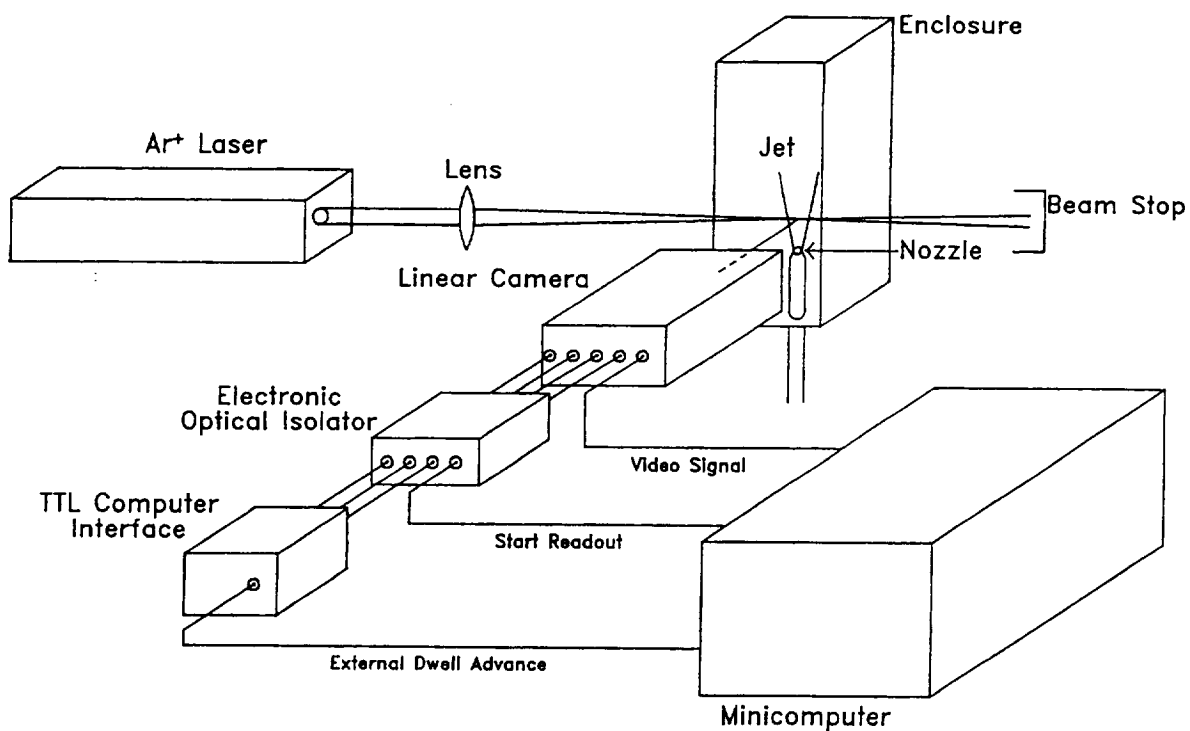


### Near-Field Concentration / Mixing Enhancement



## Photograph of laser beam in Rayleigh Light Scattering Facility





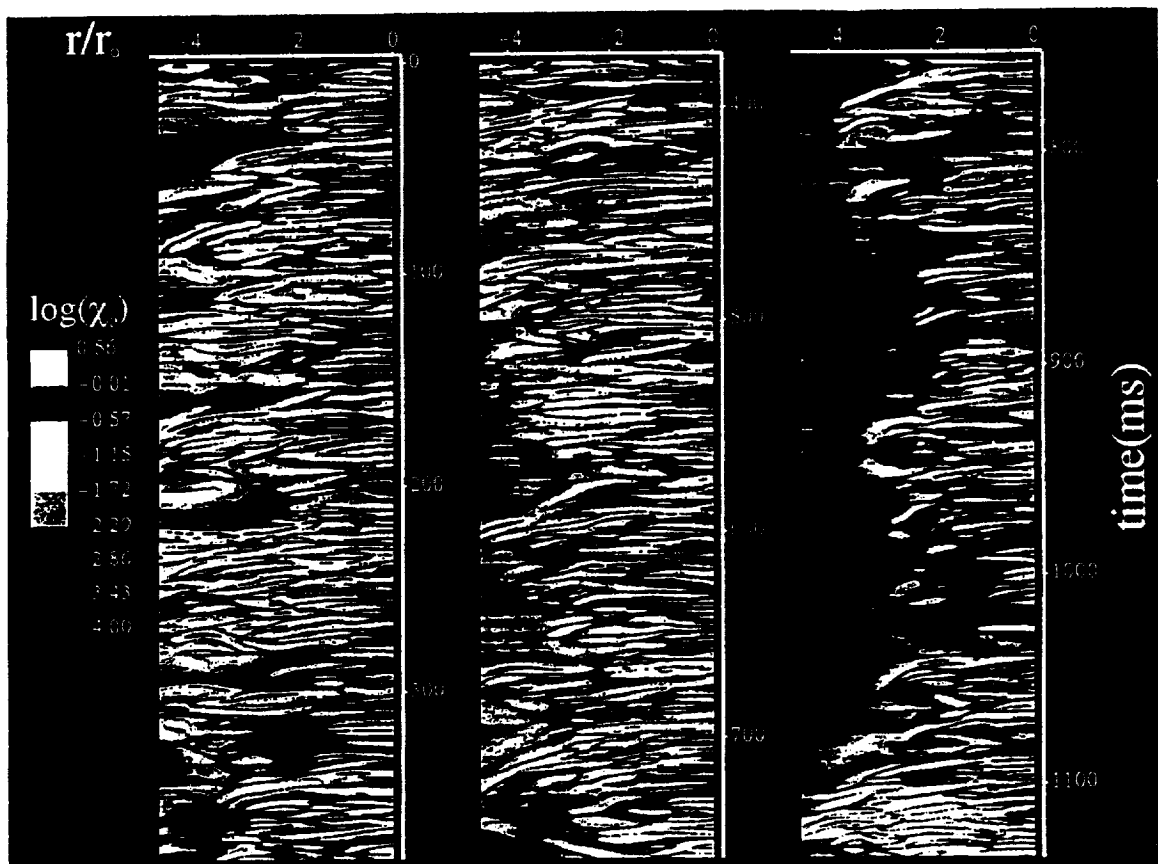
## SCALAR DISSIPATION

- Rate of molecular mixing

$$\chi = \chi_z + \chi_r + \chi_a = 2D \left[ \left( \frac{\partial \xi}{\partial z} \right)^2 + \left( \frac{\partial \xi}{\partial r} \right)^2 + \left( \frac{\partial \xi}{\partial a} \right)^2 \right]$$

Widely studied to understand:

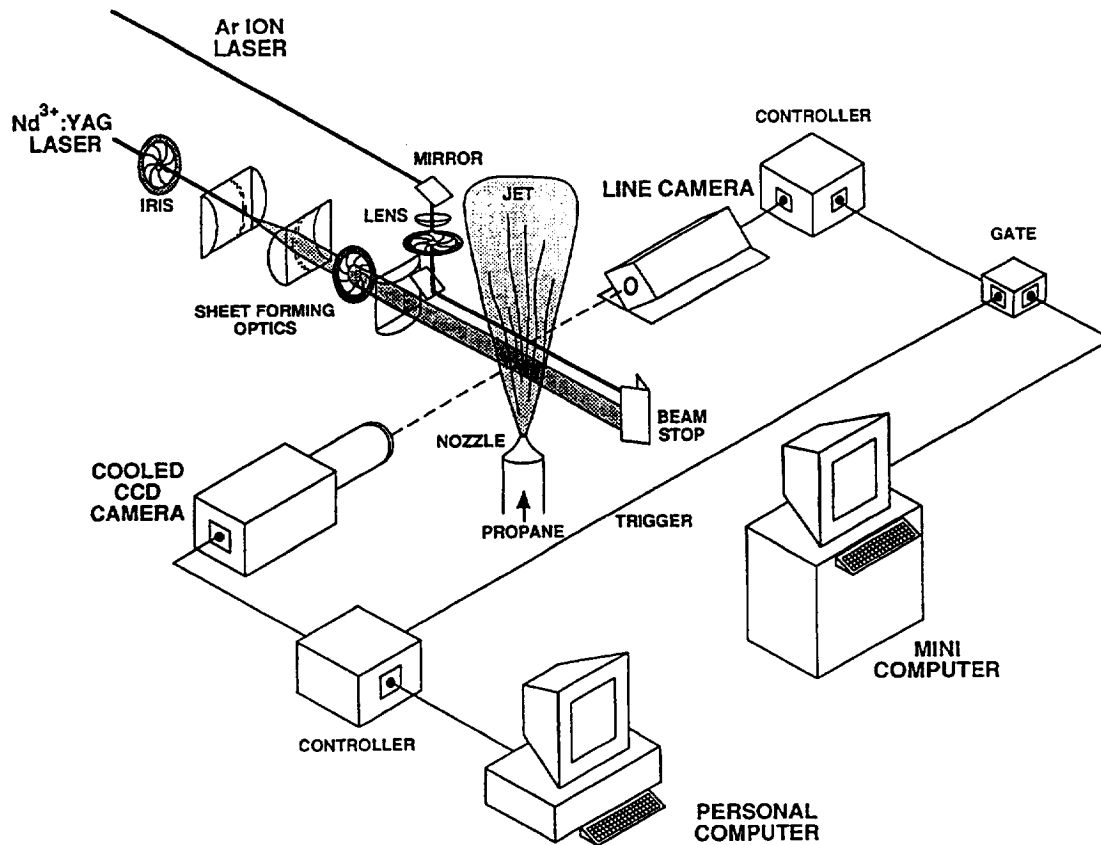
- 1) Isotropy of mixing in turbulent flows
- 2) Chemistry/turbulence interactions in turbulent diffusion flames



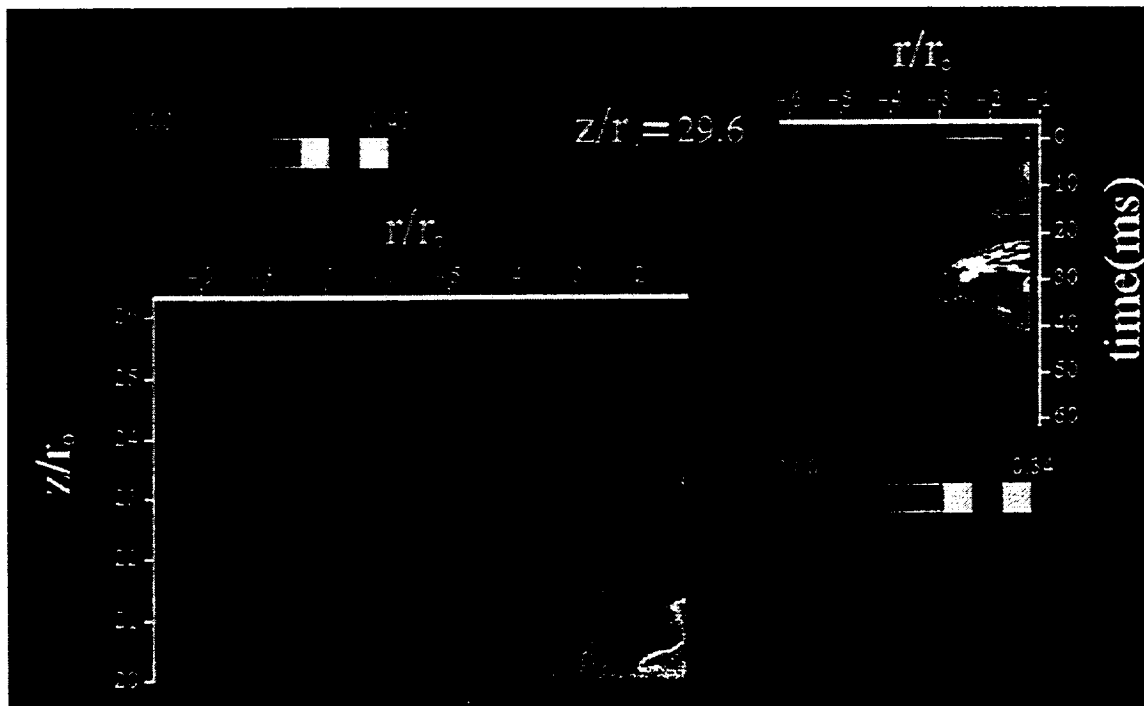


## Photograph of two-dimensional slice of axisymmetric jet using Mie scattering

## Photograph of skier, baseball player, and football player taken with a line camera



# **Photograph of $\text{Nd}^{+3}/\text{YAG}$ two-dimensional laser sheet and Ar ion laser beam**



## SUMMARY

- Experimental Systems
  - Rayleigh Light Scattering Facility
  - Real-time line camera
  - Combined instantaneous two-dimensional and line camera imaging of concentration
- Measurements discussed
  - Similarity of variable density flows
  - Shear-layer structure and side jets in absolutely unstable jets
  - Large-scale turbulent structures and scalar dissipation
- Current Efforts
  - Measurements in a helium buoyant jet
  - Simultaneous point measurements of concentration and two components of velocity in propane and methane jets

